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EFFECT OF Nb ELEMENTCONTENT IN U-Zr ALLOY ON HARDNESS, MICROSTRUCTURE AND PHASE FORMATION

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ABSTRAK

EFFECT OF Nb ELEMENTCONTENT IN U-Zr-Nb ALLOY ONHARDNESS, MICRO-STRUCTURE AND PHASEFORMATION. Experiments to determine the effect of Nb element in the U-Zr alloys on hardness, microstructure and phase formation has been done. The addition of Nb element would effect the hardness, microstructure and phase which formed. The U-Zr-Nb alloy was made with the variation of Nb 2%, 5% and 8% by melting in an electric arc melting furnace that equipped with water cooling and the argon atmosphere. The U-Zr-Nb alloy to be cut divided to some testing, such as hardness test, microstructure, and phase analysis. Hardness testing was done by Vickers hardness testing equipment, microstructure by an optical microscope, and diffraction pattern by XRD and phase analysis was done by GSAS. Hardness testing results showed that the addition of 2% to 5% Nb elementin U-Zr alloys will increased in hardness, but the addition of Nb element over 5% the hardness was decreased. Observations the microstructure showed that the addition of 2% to 5%Nb element, grains were formed from fine into coarse. Phase analysis for diffraction pattern showed that the phase changedfrom αU and γU (Zr,Nb)at 2% Nb to be αU, γU (Zr,Nb) and δ1 (UZr_2) phase at 5% and 8%Nb. Phase changes was followed by changes in its compositions.The composition of αU at 2% Nb was 40% increased to 81% at 5% Nb and decreased to 3.9% at 8% Nb. The composition of γU decreased from 59,86% to 14,91% with increased Nb from 2% to 5% and further increased to 52,74% at 8%Nb.

Keywords : Nb element, U-Zr alloy, hardness, microstructure, and phase analysis.

ABSTRACT

PENGARUH KADARUNSUR Nb PADA PADUAN U-Zr-Nb TERHADAP SIFAT MEKANIK, MIKROSTRUKTUR DAN PEMBENTUKAN FASA. *Percobaan untuk mengetahui pengaruh kadar Nb pada paduan U-Zr-Nb terhadap sifat mekanik, mikrostruktur dan pembentukan fasa telah dilakukan. Penambahan unsur Nb diduga akan mempengaruhi sifat mekanik, mikrosruktur, ketahanan korosi dan fasa yang terbentuk. Penambahan unsur Nb ke dalam paduan U-Zr dimaksudkan untuk memperluas daerah fasa gamma yang stabil dan memperbaiki ketahanan korosi. Mula-mula dibuat paduan U-Zr-Nb pada variasi Nb 2%, 5% dan 8% dengan cara melebur di dalam tungku peleburan busur listrik yang berpendingin air dan dalam media gas argon. Paduan U-Zr-Nb yang terbentuk dipotong-potong untuk dikenai beberapa pengujian, diantaranya uji mekanik (kekerasan), mikrostrutur, dan fasa yang terbentuk. Pengujian kekerasan dilakukan dengan menggunakan peralatan uji kekerasan Vickers, mikrostruktur dengan menggunakan mikroskop optik, dan fasa dengan menggunakan XRD dan analisis fasa menggunakan program GSAS. . Hasil pengujian kekerasan menunjukkan bahwa penambahan Nb pada paduan U-Zr dari 2% menjadi 5% mengakibatkan kenaikan kekerasan, tetapi kemudian kekerasan turun pada penambahan di atas 5% Nb. Pengamatan mikrostruktur yang terbentuk memperlihatkan bahwa pada penambahan unsur Nb sebesar dari 2% menjadi 5% maka terbentuk butir dari kasar menjadi halus. Dilihat dari pembentukan fasa, terjadi perubahan yakni dari αU dan γU pada 2% Nb menjadi fasa αU,γU dan δ1 (UZr2) pada 5% Nb dan 8% Nb. Perubahan fasa tersebut juga diikuti dengan perubahan prosentase fasa yaitu pada 2%Nb, dimana fase αU yang semula 40% mengalami kenaikan menjadi 81% pada 5% Nb dan selanjutnya turun menjadi 3,9% pada 8% Nb. Untuk fasa γU terjadi penurunanyang semula 59% menjadi 14% pada 5% Nb dan selanjutnya naik menjadi 32 % pada 8% .*

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Katakunci *:Logam Nb, U-Zr,sifat mekanik, mikrostruktur, fasa*.

INTRODUCTION

Development of nuclear fuel for research reactor wasto achieve high density fuel, in that case the fuel had higher mass uranium in per unit volume of the fuel.Pure uranium metal used as nuclear fuel had disadvantage in fabrication process to nuclear fuel element as final product.Pure uranium should alloyed with another metals to improve its properties. Metals such as Al, Si, Mo, Zr, Nb, Ti are commonly used as alloying metal to improve the properties of uranium metal[1]. Besides in the form of metal alloys, the uranium fuel also in the form of oxide U_3O_8 and nitride(U-N). Alloys U-Alx/Al, oxide U_3O_8/Al and metal alloys U_3Si_2/Al has been used, but they had low density of around 2,3gU/cm^3 for U-Alx / Al;3.2 gU/cm^3 for U_3O_8 /Al and U_3Si_2/Al for 4,8 gU/cm^3[2].Based on these facts, the Argonne National Laboratory on behalf of the RERTR Programme presented, by the end of 1996, an irradiation plan for new fuels allowing theoretically to reach densities up to 8 or 9 g Ut / cm^3. The Uranium alloys used for these fuels were supposed to be a γ stabilised phase. For really higher densities compared to U_3Si_2, uranium molybdenum (U-Mo) and uranium niobium zirconium (U-Zr-Nb) alloys were mainly considered[3]. Hence it is necessary obtaining higher density nuclear fuel.At the present, effort to develope the nuclear fuel with high density still continues with research and development alloys U-Mo, U-Zr and UN as nuclear fuel.

Besides the U-Mo alloy and the U-N, the U-Zr alloy was selected as the candidates of nuclear fuel that had high density. Selection the U-Zr alloy was based on several reasons, including: have a low neutron absorption cross section, have a fairly high density, have good corrosion behavior. Development of the U-Zr alloys has been done by varying the Zr element at 2%, 6%, 10%, and 14% and has been characterized and selected to be made into a mini fuel element plate (FEP)[3]. Manufacturing and characterization on the U-Zr mini FEP at 6%Zr (U-6Zr) showed acceptable results and meet the require-ments as nuclear fuel. Further prepared for irradiation test in nuclaer reactor using U enrichment at 19.75%[3]. Research manufac-ture of fuel alloy U- 4Zr- 2Nb with density of 3.17 g / cm^3 has been done, and was based on the irradiation test, neutronic properties, and the fabrication was found that the alloy is good enough as a fuel when the density of the alloy reaches 8,0 g / cm^3[4,5].

In the reactor, the nuclear fuel will undergo the changes as a result of conditions the reactor (the environmental) that affect it's performance in the reactor. To improve the performance of U-Zr alloys fuel, it is necessary to add another metal element. Metallic elements commonly added were Mo, Nb, Zr, Ti[6]. Bruno, et.al have made U-Zr-Nb alloy powder with hydriding-dehydriding method and passivated, resulting U-Zr-Nb powder with anguler morphology and irregular shape[7]. An other experiment that observed corrosion on U2.5Zr7.5Nb alloy was conducted by Celiade Afigueiredo et.al. Heating up to 300 ^oC showed good corrosion resistance, meanwhile, heating up to 600 ^oC showed lower corrosion resistance. This behavior can be associated to the phase stability when transition elements were added to uranium[8,9]. In ternary phase diagram system, resulted from experiments conducted by Dwight and Mueller can be shown that the phase γU (UZrNb) will be stable for in the range where the phase δ (UZr2) were existence at temperatures higher than 700ºC. The solubility of Nb in phase δ(UZr2) is about 10% by weight. Phase γU (Zr, Nb) can be maintained as metastable phase depends on the cooling rate and the concentration of Nb and Zr[10].Addition of Nb influence the properties of U-Zr alloy, such as mechanical properties, microstructureand formed phases.Addition of Nb in specific limit will increased mechanical properties such as it's hardness. Addition of Nb more than it should, caused increased it's hardness and made it harder to process should avoided.Alloy hardness correspon-ded to it's microstructure such as grain size and phases.Type and number of phases besides influence the mechanical properties also influence the alloy stability at high temperature.This research aimed to make theU-Zr-Nb alloy on the variation of Nb and to understand the changes on it's properties.

METODOLOGI

Uranium, zirconium, and niobium metals which has been cut into pieces placed in the electric arc furnace with argon atmospheric and cooled water to be melting. Alloying metals ofU, Zr, and Nb was done with varying the Nb metal, respectively for 2, 5 and 8% by weight while the Zr metal fixed at 6%. Each pair of samples U, Zr and Nb was melted with electric current at 150 A and remelted for five times which each side done for 3 minutes to ensure achieved homogeneous ingot U-Zr-Nb alloy. The total weight of ingot U-Zr-Nb for one smelting was 10 g. Similarly to the other pairs of samples of U-Zr-Nb done in the same way. The ingots the U-Zr-Nb alloy then cut into pieces to be subject to testing, such as: hardness, microstructure and the phase are formed. Hardness testing is done by using a micro hardness test equipment (Vickers), micro-structure by using an Nikon Ephiphot optical microscope, and phase using Panalytical Empyrean X-ray Diffraction (XRD). The phase analysis done by GSAS software from diffraction pattern obtained from X-Ray Diffraction (XRD). The total weight of ingot the U-Zr-Nb for one smelting is 10 g. Similarly, for a pairs of samples of U- Zr-Nb the other was done in the same way.

RESULT AND DISCUSSION

a. HARDNESS

Hardness result shown in Figure 1, the microstructure in Figure 2.a, b and c, and the results of testing the phase is shown in Figure 3.a, b and c.

Figure 1 shown the data hardness test, at the higher Nb content of up to 5% by weight, then the higher the hardness of 375 into 523 HVN, then decreased when the content of Nb rise at 8% Nb alloy.The U- 6Zr- 8Nb hardness at 319 HVN.This condition is caused by the addition of Nb element into the U-Zr alloys which form the U-Zr-Nb alloy.During melting Nb elements form nucleus, and these nucleus would growth become grains.The Nb element at 2% relatively remain small to solubility limit compared at 5%.Nucleations at 2% fewer than 5% and resulting fewergrains formed. Grain act as an obstacle to dislocation motion, more grains are formed, the movement of dislocations will be more difficult and lead increase hardness. Therefore, the Nb addition at 5% into U-Zr-Nb alloy leadthe hardness ofU-Zr-Nb alloy higher than the addition of 2% Nb.The addition of Nb elements exceed 5% showed hardness decreasing. The hardness decre-asing caused by the abundance of Nb elements in alloys and will slow the formation of grain from the nucleus of the Nb. In other words, the grain growth rate is slower than the speed ofthe nucleation rate for Nb element addition. Therefore increased Nb above 5%, caused the grains fewer. Formed grains fewer caused the movement of dislocations becomes easier and decreased in hardness.

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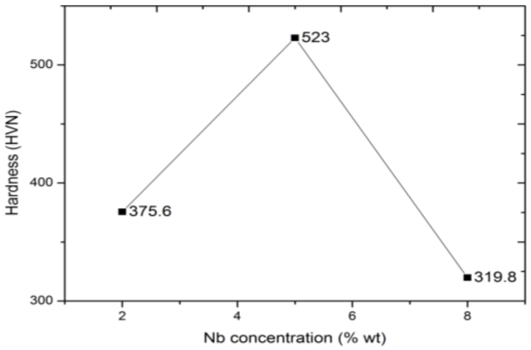
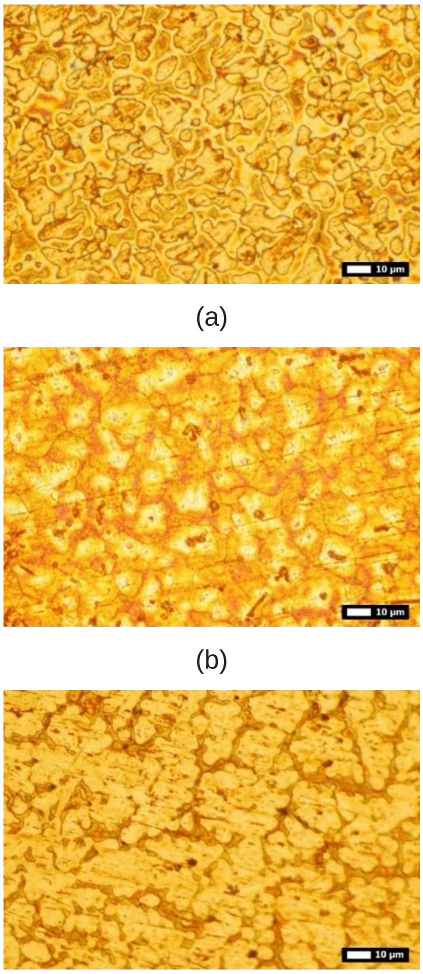


Figure 1. Curve of hardness testing versus percentage of Nb content

b. MICROSTRUCTURES

Figure2.a, b and c shows the microstructure images of U-Zr-Nb alloy for the addition at 2% (Figure 2.a) formed grains are not uniform, partly in the form of coarse grains and partly others fine-grained. The grain structure showed the alloy not uniform during melting process. These was caused in arc melting process limited the movement of molten elements during heating.Repetition five times heating with arc melting was done to increased its homogenous compare to once heating. Compared to the U-Zr alloys with Nb addition at 5%(U-6Zr-5Nb) as shown in Figure2.b, it's seen that the alloy of U- 6Zr- 5Nb have finer grain size than the alloy ofU-6Zr-2Nb (Figure2.a).This is due to the amount of Nb element is added to thealloyU-6Zr-5Nb more than Nb element is added to the U-Zr alloys with the addition at 2% (U- 6Zr- 2Nb), caused more nucleus were formed if the nucleus were formed higher then the grains were formed higher and finer. However, on the Nb addition at 8% which is slightly near to the solubility limit of Nb in U- Zr alloy also show less grains and more coarse than Nb at 2% and 5%.TheNb concentration near it's solubility limit in U-Zr alloy only some Nb can soluble and continues to nucleation and grain growth with U-Zr alloy. The othersmall amount of Nb that can not soluble in U and Zr would acted as grain growth restrain of U-Zr-Nb alloy.The grain size value, on the Nbaddition at 2%, 5% and 8% respectively U-Zr-Nb alloy had an average grain size at 0,012 µm; 0.0226 µm and 0,013µm or with range from (0.009 to 0.025) µm; (0.013 to 0.029) µm and (0.006 to 0.02) µm respectively.



(c)

Figure 2. Microstructure images of U-Zr-Nb alloys.
a. U-6Zr-2Nb,
b. U-6Zr-5Nb, and
c. U-6Zr-8Nb

c. PHASE ANALYSIS

Powder diffraction pattern showed in Figure 3a,b and c. Figure 3.a is diffraction pattern for U-6Zr-2Nb alloy which indicates that the alloy U-6Zr-2Nb have αU and γU phases. The αU phase appears on the 2θ angles respectively at 35.4 °; 38.6 °; 52.05 °, and 65.01 ° with each hkl of 021, 111, 200, and 211, while the phase angle γU appears on the 2θ of 35.64 °; 59.8 °, and 76 ° with each hkl of 002, 200, 131, and 220. The αU phase has orthorhombic structure in space group of Cmcm with lattice parameters a = 2.917 (5) Å, b = 5.82 (1) Å, and c = 5.02 (1) Å, withdensity at 18.428 g/cm^3. Weight percentage composition of existing αU phase at 40.14%. The γU phase has a cubic crystal structure in the space group of Im-3m with lattice parameters a = 3.497 Å, density at 18.431 g/cm^3. Weight percentage composition of existing γU phase at 59.86%. The U-6Zr-5Nb alloy consists of three phases, namely phase αU, γU and δ1 (UZr2). The αU and γU phases appears at an angle 2θ and hkl that the same as in theU-6Zr-2Nb alloy. Similarly, the lattice parameters and thespace group αU and γU phases also the same as the alloy U-6Zr-2Nb. The αU and γU phases density respectively are at 18.127 g/cm^3 and 18.657 g/cm^3, while the weight percentage compositions of αU and γU phases respectively are at 81.80% and 14.91%. Meanwhile, the phase δ1 (UZr2) appears withstrong peak at 2θ angle of 26.67 ° and the hkl 001.The δ1 (UZr2) phase has a hexagonal crystal structure with lattice parameters of a = 6.529(2) Å and b = 3.61(3) Å ,and density of 9.754 g/cm^3. Weight percentage composition of δ1 (UZr2) phase at 3.29%.

TheU-6Zr-8Nb alloy consists of three phases, namely phase αU, γU and δ1 (UZr2).The αU and γU phases appears at an angle 2θ and hkl the same as in the alloy U-6Zr-2Nb and U-6Zr-5Nb. Similarly, the lattice parameters and the space group αU and γU phases also the same as the alloy lattice parameter and the space group on the alloy U-6Zr-2Nb andU-6Zr-5Nb. The αU and γU phases density respectively are at 20.067 g/cm^3 and 19.909 g/cm^3, while the wightpercentage compositions of αU and γU phases respectively are at 3.10% and 52.74%. Meanwhile, the δ1 (UZr2) phase appears at 2θ angle of 26.67 ° andhkl 001. The δ1 (UZr2) phasehas a hexagonal crystal structure with lattice parameters a = 6.529 Åand b = 3.61 (3) Å,well as density of9.607 g/cm^3. Weight percentage composition of δ1 (UZr2) phaseat 44.16%. The results of this experiment corresponds to an experiment that conducted by Komar Varela, et al (2012), in which the experiment to make U-Zr-Nb alloys obtained of αU, γU, and δ1 (UZr2) phases[9]. The test results of XRD shown that the increase in the Nb contents shows changes in phases content that αU and γU at 2% Nb into αU, γU and δ1 (UZr2) phases at 5% and 8% Nb. The phase changes was followed by the weight percentage composition change in the phase at 2% Nb, αU phase which was originally 40% increased to 81% at 5% Nb and further decreased to 3.9% at 8% Nb.The γU phase decrease which was originally 59 % to 14% at 5% Nb and further increased to 32% at 8% Nb. Meanwhile, the δ1(UZr2) phase found in 5% Nb increased in 8% to 44% Nb. At Figure 4 it was showed the curve correlation beetwen Nb concentration versus phase concentration (α, γ dan δ).

At 5% Nb, the UZrNb alloy hardness is higher than the 2% and 8% Nb. This is due to 5% Nb the U-Zr Nb alloy containing alpha-phase highest compared to 2% Nb alloy and 8% Nb. The phases in three alloys are αU,γU, and δ. Note that the alpha phase having a crystal structure orthorombic, whileγ U and δ phases cubic and hexagonal respectively. At 2% Nb, the delta phase is very low to be detected by XRD or not formed and tend to form alpha and gamma phase. Alpha phase is due during the cooling process when the change in the gamma phase into alpha phase[11]. Increasing concentrations of Nb to 5% to encourage the formation of delta phase and by Ghosal, et.al (2014) Nb concentrations of more than 3.5% will inhibit the formation of beta phase. It also results in a high concentration of alpha phase in this alloy which occurs due to the gamma phase change that occurs as a result of negligent phase during the cooling process. The increase in Nb concentration to 8% that

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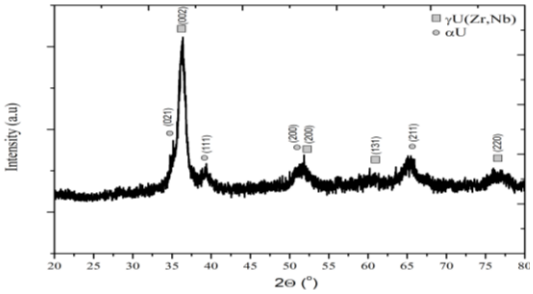
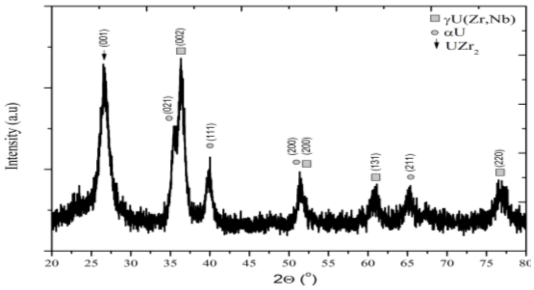
encourage the formation of more stable gamma phase[11]. This is because the temperature for the formation of gamma phase will be lower due to increased concentrations of Nb[11]. In the phase diagram of U-Nb and U-Zr shown that increasing Nb concentration will also increase the amount ofdelta phase formed[11].



(a)



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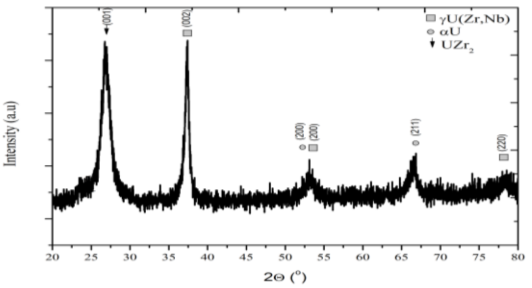

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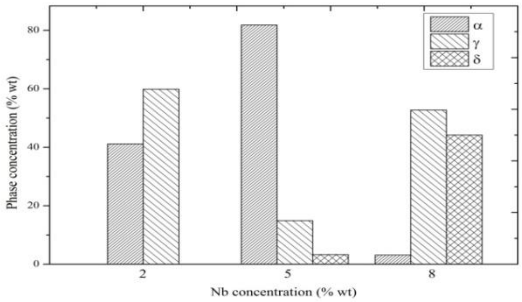
Figure 3. Diffraction pattern of U-Zr-Nb alloys
(a) U-6Zr-2Nb,
(b) U-6Zr-5Nb and
(c) U-6Zr-8Nb



Figur 4. Curva of Nb cocentration versus phase concentration

CONCLUSIONS

The results of hardness testing, microstructure and phase analysis of the U-Zr-Nb alloy can be concluded that the addition of Nb element into the U-Zr alloys will increase the hardness of up to 5% of Nb content, then decreased at higher Nb element. The Nb element addition at 2% and 5% shows changes in the microstructure which increasingly fine grain size, but the addition above 5% the grains become coarse (enlarged). Phase changes occur on the addition of the Nb content of αU and γU at 2% Nb into phase αU, γU and δ1 (UZr2) at 5% Nb and 8%Nb. The phase change concentration was followed by content of phase. At 2% Nb, the the concentration αU increased from 40% to 80 %. γU phase at 2%Nb decreased from 59,86% down to 14,91% and at 5% Nb and increased to 52,74% at 8% Nb.

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